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A UNIFIED THEORY OF 1/F NOISE AND DIELECTRIC RESPONSE IN CONDENS--ETC(U)

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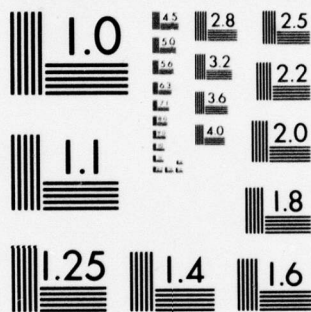
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10 K. L. NGAI

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20. Abstract (Continued)

We have thus a unified theory explaining on the same basis both physical phenomena with a single physical picture.

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A UNIFIED THEORY OF $1/f$ NOISE AND DIELECTRIC RESPONSE IN CONDENSED MATTER

I. Introduction

When a current is passed through a material, spontaneous fluctuations in the current and/or voltage are invariably present and this phenomenon is commonly referred to as "noise." There are various causes for fluctuations and noises.¹⁻³ Among them are thermal noise and shot noise, and both are by now well understood. However, in almost all current-carrying condensed matter (e.g. semiconductors, insulators, metals, liquid electrolytes, ionic conductors, biopolymers, etc.) and in all solid state devices (e.g. MOS transistors, infrared detectors, microphones, amplifiers, etc.) a considerable low frequency noise is generated over and above thermal noise and shot noise.¹⁻⁴ The predominant noise component at low frequencies has a characteristic power spectral density $G(f)$ given approximately by

$$G(f) \propto I^2/f^\beta \quad (1)$$

where I is the direct current flow, f is the frequency, and β is close to unity. This low frequency additional noise is generally known as flicker noise, $1/f$ noise or excess noise. In Eq. (1), the power spectrum or spectral density $G(f)$ of the fluctuating current $\delta I(t)$ describing a stationary random or stochastic process is defined as the ensemble average of the time average of the power dissipation in unit resistance per unit frequency bandwidth. This definition can be equivalently restated as an identity^{2,5}

Note: Manuscript submitted November 29, 1978

$$\overline{\langle \delta I^2(t) \rangle} = \int_0^{\infty} G(f) df \quad (2)$$

where $\overline{\langle \delta I^2(t) \rangle}$ is the time independent ensemble averaged mean square current fluctuation.

1/f noise is an ubiquitous phenomena. As a rule it is present in any current-carrying material and/or device and any experimenter is bound to reveal its presence in his system if he measures the noise to low enough frequencies. Once 1/f noise has appeared it seems to persist with its approximately 1/f dependence indefinitely as we go to lower and lower frequencies. Thus 1/f noise is a fascinating universal phenomenon of nature that at the same time has important implications and consequences on electronic device operations and the stability of the phase and frequency of high-frequency generators. Numerous attempts have been made to unveil the fundamental mechanism of the 1/f noise in various systems but it is generally agreed upon, as expressed in a recent review,⁴ that it remains as an unsolved problem.

The present paper is yet another attempt to understand the fundamental nature of the 1/f noise. The present author certainly agrees, as has been remarked,⁴ that "in the field of 1/f noise we have seen a long tradition of models being proposed that are forgotten after a short time. The physics they are based upon is either too special or too artificial . . ." Heeding this remark and in the course of bringing ourselves to write this article, we have, as much as we can, convinced ourselves that in our model to be proposed the physics involved is neither too special nor artificial and our model has a good chance of not being forgotten in a short time. The reason for this belief is

that, as we shall see, the physics involved in our model is manifestly general and universal. It is also a unified theory that attributes both the ubiquitous $1/f$ noise and the "universal" dielectric response to originate from the same mechanism, and further trace both sources to the correlated state excitations. The meaning of the "universal" dielectric response and the identity and nature of the correlated state excitations have been expounded in our earlier treatment⁶⁻⁸ of dielectric response of condensed matter in general, and shall be restated here in a condensed form in Section II. Briefly here the term "universal" dielectric response stands for another ubiquitous low frequency behavior of both the real and imaginary parts, $\epsilon'(\omega)$ and $\epsilon''(\omega)$ respectively for dielectrics that have current-carrying charged species (electrons, holes or ions) in them. Then invariably measurements of the dielectric response at sufficiently low frequency will uncover that both ϵ' and ϵ'' have the characteristic frequency dependence of ω^{n-1} , with n positive but small. At the mean time we return to emphasize our unified theory distinguishes itself from all earlier models in pointing out for the first time that the low frequency dielectric response and the $1/f$ noise are both universal behaviors that will show up simultaneously in any current-carrying material or solid state device, and they are implicated to have the same physical origin. In this sense our approach is not superficial but rather tends to be profound; and not artificial because it links the $1/f$ noise together with another universal phenomenon to fundamental structure and excitations of matter. As we shall see in later sections that our mechanism works in the bulk as well as at interfaces and hence our theory can reconcile the controversy of whether $1/f$ noise

is a surface or bulk effect. Another issue of current interest is whether $1/f$ noise stems from mobility fluctuations or from number fluctuations. Our approach is able to derive $1/f$ noise from mobility fluctuations. In a way our model has adequately considered mobility fluctuations, while no other models that have been proposed have considered such mobility fluctuations.⁴ The organization of the paper is as follows. Section II brings out the universal nature of the dielectric response of current-carrying materials at very low frequencies, summarizes the theory we have developed very recently⁶⁻⁸ for its explanation, and restate the basic physical ingredients of the model. These same basic physical ingredients are then reemployed to derive the $1/f$ noise power spectrum in Section III. Section IV deals with the predictions, consequences and generalities based on the results of this model, and compares/contrasts our approach with others. A brief summary is given in the last Section V.

II. Universal Dielectric Response: Infrared Divergent Response by Excitations of Correlated States

The dielectric response of solids and liquids has been the subject of intense investigation over a long period of time extending to this date, and are pursued by physicists, chemists and engineers alike. Jonscher⁹ has studied and evaluated a vast amount of experimental data on a wide range of solids that have led him to the formulation of a general classification of all types of dielectric responses below 10 GHz which are common to materials of widely different physical, chemical, structural and geometrical properties. The various types of dielectric response are summarized in Fig. 1. It was observed⁹ that the dielectric

response functions in frequency or in time depart strongly from the Debye response and fall into a remarkably common or universal pattern. The outstanding feature is that the frequency dependence of the dielectric loss $\epsilon''(\omega)$ (i.e. the imaginary part of the dielectric function shown in Fig. 1 as the solid line in $\log \epsilon''$ vs $\log \omega$ plots) follows the empirical law

$$\epsilon''(\omega) \propto \omega^{n-1} \quad (3)$$

with $0 < n < 1$, extending over many decades of frequency. Empirical law (3) when obeyed exemplifies as a straight line segment in the $\log \epsilon''$ versus $\log \omega$ plots as in Fig. 1, and the common occurrence of this behavior can be inferred by inspection. For dielectrics with permanent dipoles, a broad loss peak occurs and at low enough frequencies the empirical law (3) ceases to be valid. For dielectrics that contain current-carrying species that can give rise to dc conductivity, independent on whether the dielectric is dipolar or not and whether a loss peak shows up or not, it is common when going down in frequency the empirical power law (3) will start to hold at some low frequency value and continue to do so down to the lowest available frequencies. For nondipolar dielectrics without any loss peak present in $\epsilon''(\omega)$, quite often in the entire frequency domain^{7,9} the response is a superposition of two such empirical power laws:

$$\epsilon''(\omega) \propto (\omega/\omega_c)^{n_1-1} + (\omega/\omega_c)^{n_2-1} \quad (4)$$

with n_1 and n_2 representing the two slopes in the plot on the extreme left and shaded portion of Fig. 1, and ω_c is the characteristic frequency at which the transition from one power law to the other occurs. For $\omega \ll \omega_c$, that power law term in Eq. (4) with the smaller value of n

dominates. It is generally true that the smaller n has small positive value no greater than 0.3, thus the form of low-frequency response corresponds to both ϵ'' and ϵ' rising as ω^{n-1} steeply towards low frequencies, as shown in the darkest shaded portion of Fig. 1. This dielectric behavior, a special case of the more universal law Eq. (4), is what we shall only need to consider in any further reference to universal dielectric response in this present work on $1/f$ noise. This is easily understandable because we are interested in a unified view of dielectric response and $1/f$ noise. Thus we are interested in both the dielectric response and the $1/f$ noise of the same material at the same frequency region. And recalling that $1/f$ noise occurs in current-carrying materials which have mobile charged carriers and at the lowest frequency, it is clear that our attention can be confined to only the darkest shaded panel of Fig. 1 which will be dubbed " $1/\omega^{1-n}$ dielectric response."

The $1/\omega^{1-n}$ dielectric response is ubiquitous and appears in various materials with mobile charged carriers including inorganic ceramics, ionic conductors, polymeric materials, inorganic crystalline and amorphous materials, insulating or semiconducting, and organic and biological systems. It is valid in covalent, ionic and molecular solids, in single crystals, polycrystalline and amorphous structures. These hallmarks for the $1/\omega^{1-n}$ dielectric response are remarkably similar to those of $1/f$ noise. Dielectric response and fluctuation can hardly be considered as disjointed or unrelated physical phenomena. It is reasonable to demand any theory proposed to explain the origin of the $1/f$ noise should account for the $1/\omega^{1-n}$ dielectric response simultaneously, and vice versa. Any theoretical treatment of the fundamental

aspects of one universal phenomenon should be extended to cover the other universal phenomenon for consistency, for completeness and for credibility.

We have recently proposed a fundamental mechanism for the $1/\omega^{1-n}$ dielectric response. In a broad classification of dielectrics according to the type of interaction or correlation inherent in different material we have found always, independent of the bonding, structure, interactions, current-carrying species etc., the presence of one type of "correlated states" or the other. Low energy excitations of these correlated states with excitation energy E has energy density of excitations $N(E)$ (defined conventionally as the partial derivative of the total number $N_{\text{total}}(E)$ of the correlated state excitations with excitation energies less than E with respect to E , i.e., $N(E) = \partial N_{\text{total}}(E) / \partial E$) proportional to E . The charged particles responsible for dielectric polarization undergo quantum transitions, changing their positions between preferred sites by hopping or jumping movements. The times taken by these transitions are negligible compared with the time required for excitation of the correlated states. This follows from the nature of the correlated states. Hence, as far as the correlated states are concerned, the charged particles jump spontaneously. A sudden change of the potential induced by the charged particle on the correlated states takes place. The transient response of the dielectric to the sudden switch on of a new potential involves the emission of low-energy excitations of the correlated states. At long time, the transient responses via Fourier transform is essentially the low-frequency dielectric response.⁶⁻⁸ The sudden potential change V does not depend on the correlated state

excitation energy E . Then,

$$V^2 N(E) = nE \quad (5)$$

is proportional to E , and the condition for infrared divergent response⁷⁻¹¹ of the correlated states is satisfied. Infrared divergent response means that there is an increasingly high probability of exciting decreasingly small energy correlated state excitations, and cause the power law $1/\omega^{1-n}$ divergence in the response. Whenever the condition $V^2 N(E) = nE$ is satisfied, the number $p(E)$ of correlated state excitations emitted with energy $E = \hbar\omega$ per unit energy interval is proportional to $1/E$; the energy \bar{E} emitted in correlated state excitations per unit energy interval is constant; and the total number p_{tot} of correlated state excitations emitted in the transient response is weakly (logarithmically) divergent. These statements follow straightforwardly from Eq. (5) and quantum mechanical perturbation theory which gives the following expressions for $p(E)$, \bar{E} and p_{tot} ,

$$p(E) = V^2 N(E)/E^2 \quad (6)$$

$$\bar{E} = E p(E) \quad (7)$$

$$p_{\text{tot}} = \int_0^{E_c} V^2 N(E) dE/E^2 \quad (8)$$

where the upper cut-off energy E_c may be chosen as the energy region over which Eq. (5) ceases to be a good approximation.

The nature of the correlated states are intriguing, may differ greatly dependent on the structure, bonding, charged species and/or dipoles but invariably have the common property that they will satisfy the condition for infrared divergence. For diamagnetic dielectrics with electronic interactions and correlations need to be considered, electron spin up-spin down pairing interaction leads to strongly

self-trapped (due to bond, atomic or molecular distortions) correlated states which are now localized paired states. The majority of dielectrics in nature is diamagnetic and, in part, this can be attributed to actions of closed shells, lone pairs, covalent bonding and orbital hybridizations. It is in this class of diamagnetic dielectrics that strongly localized electron pair states comprise one type of correlated states that give rise to infrared divergence as has been discussed in detail in our treatment.⁶⁻⁸ Similarly strongly self-trapped single electron states in dielectrics with electronic paramagnetism are another source of correlated states that will also have an infrared divergent response. In dielectrics such as electrolytes, ionic conductors, polymeric solids etc., where molecular, ionic and dipolar interactions are apparent there is another class of correlated states of interest. Here a correlated state specified by some generalized coordinates of a set of molecules, atoms, ions or dipoles correspond to a local energy minimum. Low energy E excitation of one correlated state to another can be shown, by very general arguments,^{7,8} to have density of excitations $N(E) \propto E$, i.e., proportional to E . Considerations of the transient response of these correlated states to charged carrier transport motion lead again to infrared divergent response. Infrared divergent response of the correlated states leads to dielectric loss $\epsilon''(\omega)$ to have the frequency dependence of ω^{n-1} exactly as in Eq. (3) with n given by Eq. (5).

We have stated only the essence of our theory of the $1/\omega^{1-n}$ universal dielectric response. The fundamental point is that independent of the dielectric type, some correlated states will invariably be present that dominate the low-frequency response. Since details of our

theory have been documented elsewhere, we have given here a skeleton of the physical ideas in the theory. The predictions of the theory do not end at obtaining the $1/\omega^{1-n}$ law for dielectrics with charged carriers. Specializing to dielectrics with dipoles, the theory predicts a most unusual transient response function of the form at long time

$$\psi_i(t) = t^{-n} \exp(-a t^{1-n}) \quad (9)$$

The Fourier transform of Eq. (9) to frequency ω -domain gives both the real and the imaginary parts of the dielectric response function. What is remarkable is that Eq. (9) derived⁶⁻⁸ microscopically for the special case of a dipolar dielectric turns out to be an empirical response function postulated earlier by Williams and Watts¹² and pointed out by them to give remarkable fit to dielectric response data of a number of dipolar dielectrics. This happy coincidence has spurred us to analyze exhaustively the published dielectric response data. The overall agreement of the predictions of the now microscopically derived Williams and Watts empirical law is impressive.¹³ The unqualified success of the theory in dipolar dielectrics lends immense credence to infrared divergent response of correlated states to explain the low frequency dielectric behavior of matter, which is the cornerstone of our earlier dielectric theory, and is also the cornerstone of our present theory of $1/f$ noise. It is in this sense that now the $1/f$ noise and the $1/\omega^{1-n}$ dielectric response, both being ubiquitous phenomena in nature can be based on one fundamental physical concept that we call the present theory a unified theory.¹⁴

Infrared divergence in quantum electrodynamics is well known in the phenomena of bremsstrahlung or Cerenkov radiation where "soft" photons

are emitted when a high energy charged particle is scattered by nuclei.¹¹ Handel¹⁵ has taken over this infrared divergence in photon emission to explain the origin of $1/f$ noise. The common feature of our present work and Handel's is the infrared divergence principle being a fundamental ingredient in both theories. But there are large departures in the microscopic and physical ingredient between our theory and Handel's. In our approach, we have traced the origin of both the $1/f$ noise and the universal dielectric response⁶⁻⁹ to the correlated states which is fundamentally related to the structure of matter. Even with any stretch in imagination these correlated states are not the "elementary excitations" of solids as commonly known.¹⁶ In a way our earlier theory of dielectric response is important not only in it being able to explain the universal dielectric response but also in the discovery and identification of this new class of states and excitations which is an important task. Our theory unifies both the $1/f$ noise and the $1/\omega^{1-n}$ as well as the Williams and Watts empirical dielectric response phenomena.^{6-8,12} Weak (i.e. electromagnetic) interaction has the fine structure constant $\alpha = 1/137$ as effectively the coupling constant. Hence any attempt in invoking the soft photons in infrared divergent dielectric response will only obtain the $1/\omega^{1-n}$ law with $n \ll 1$, while experimentally the universal $1/\omega^{1-n}$ dielectric response has n values ranging from zero to one (Fig.1). Further it is hard to imagine that photon emission would have anything to do with the dielectric loss peak given as the Fourier transform of Eq. (9).

III. Derivation of 1/f Noise

The current conduction mechanism is varied from one type of material to another although they all exhibit the 1/f noise phenomena at low frequencies. Take for example some often quoted systems that have 1/f noise such as

(a) continuous metallic films and high purity bulk semiconductors where electron transport occurs via band conduction,¹⁷ in the process electrons are scattered by impurities, imperfections, lattice phonons, etc.;

(b) very thin metal films which have lost the bulk properties and electrical conduction is probably partially via a hopping process. Moderately doped semiconductors at low temperature where conduction occurs via electron (or hole) hopping;

(c) amorphous semiconductors or insulators where charged carriers hop from site to site, and phonons may also be involved in the hopping transition;

(d) "small polaron" molecular crystalline solid where carriers are self-trapped and transport via activation over its self-trapped induced barrier from one equivalent well to an adjacent one;¹⁸

(e) solid electrolytes and ionic crystals with ionic conductivity caused by hopping of ions from site to site¹⁹ (the solid has a network of sites where the ion can sit, and are usually more sites than ions);

(f) classical Brownian particle motion conduction systems usually describable by the Langevin equation²⁰ such as $m\ddot{x} + m\gamma \dot{x} = f(t)$ suitable for ion diffusion in liquid electrolytes, where m is the mass of the ion, γ is the damping and $f(t)$ the stochastic force caused by bombard-

ment of the ion by the molecules or other ions of the electrolyte. If the Brownian particle is moving in a periodic potential an additional term enters into the Langevin equation. For example in a one-dimensional sinusoidal potential,²¹ the corresponding Langevin equation is $m\ddot{x} + m\gamma\dot{x} + m\omega_0^2 \frac{a}{2\pi} \sin\left(\frac{2\pi}{a} x\right) = f(t)$, where $\omega_0^2 = (2\pi)^2 A/2ma^2$ with A the barrier height and a the lattice constant. It has been pointed out that the latter form of the Langevin equation should describe (i) the diffusion of ions in crystals where then x_0 assumes discrete values x_n , the different possible sites for the ions and (ii) the fluctuations of the Josephson supercurrent through a tunnel junction where then $(2\pi/a)x$ denotes the phase of the superconductive order parameter and m , $(m\gamma)^{-1}$, and $m\omega_0^2$ are the capacitance, the resistance, and the coupling energy of the Josephson junction;

(g) interface solid state devices such as MOSFET where the conduction mechanism at high density of channel carriers and moderate temperature can be subband conduction with scattering from interface charges, surface roughness, ionized impurities etc., as well as scattering into and/or out of interface states (i.e. traps). At low channel carrier densities and low temperatures, the conduction mechanism then may well be hopping over self-trapped barriers and the mobility is temperature activated.

In all these examples cited whatever the conduction mechanism, in the conduction process the charged carriers undergo transitions that will occur in time intervals much shorter than the times characteristic of the response of the correlated states⁶⁻⁸ discussed in Section II. The transitions are, for example, the phonon and impurity potential

scattering in band conduction, cases (a) and (g); and various types of hopping transitions (including phonon assisted hopping) from site to site in cases (b) to (g); and elastic and inelastic tunneling transitions in superconductive and non-superconductive tunnel junction and contacts as well as in Josephson junctions, case (f). These transitions although have time characteristics which may be long compared to elementary excitations (such as phonon, electron-hole excitations, spin waves, plasmons etc.) response time can in effect be considered as to occur in no time at all as far as the response of the correlated states are concerned. We shall use the word "sudden" henceforth in this sense to describe the carrier transition. The problem can then be considered as the transient response of the correlated states to these sudden transitions which abruptly switch on a new Hamiltonian (or new potential) acting on them. The situation is nothing extraordinary in most situations commonly considered in physics of condensed matter. An impurity in an insulating crystal can be excited from its ground electronic state to an excited electron state by the absorption of a photon.¹⁰ From the Franck-Condon principle it is clear that the electronic optical excitation can be considered as if it occurs instantaneously as far as the vibronic excitation (i.e. phonons) are concerned. The vibronic coupling of the electron in the ground state is different from that of the electron in the excited state. Hence the optical transition switch on a new Hamiltonian for the vibronic excitations to respond. The transient response of the vibronic excitations contributes normally a "side-band" to the electronic optical absorption, without essential modification of the ground to excited state transition, as commonly observed in most

spectroscopy. However, transient response is extraordinary when the conditions for infrared divergence is satisfied by the vibronic excitations, the transient response extends to and weakly divergent at arbitrary long time or low frequencies, and at the same time completely modifies the ground to excited state transition to the extent that one no longer recognizes it.¹⁰ We have seen that the correlated states of Section II do satisfy the rather stringent requirements, Eq. (5), for infrared divergent response to sudden transitions of charged carriers that will occur in the process of conduction. It is this concomitant infrared divergent response of the correlated states to charged carriers in the process of conduction that will give rise to the $1/f$ noise to be derived in the remainder of this Section.

Sudden transitions from state Φ_i to Φ_j of charge carriers in transport cause infrared divergent response of the correlated states. This is reflected in the probability distribution $P(\epsilon)$ of energy ϵ of the correlated state excitations excited in the transient response having the energy dependence of the form $1/\epsilon^{1-n}$, where n is given via Eq. (5). The exact expression for $P(\epsilon)$ can be obtained^{7,10} as

$$P(\epsilon) = (2\pi)^{-1} \int_{-\infty}^{\infty} dt \exp \left\{ i t \epsilon / \hbar \right\} e^{-\phi(t)} \quad (10)$$

where $\phi(t)$ describes the time response of the correlated states. In subsequent development we shall put $\hbar = 1$ and use ϵ and ω interchangeably. When condition expressed as Eq. (5) is satisfied, $\phi(t)$ has the form $\phi(t) = n \int_0^{\epsilon_c} d\epsilon \epsilon^{-1} \{ 1 - \exp(-i\epsilon t) \}$. At large t , $\exp(-\phi(t))$ has the form of $e^{-\gamma (\epsilon_c t)^{-n}}$, where γ is a constant of value .57221. $P(\epsilon)$ has been evaluated¹⁰ and the approximate result given as

$$P(\epsilon) = \left[\frac{1}{\pi \epsilon_c} \right] e^{-\pi \gamma} \sin(\pi n) \Gamma(1-n) \left(\frac{\epsilon_c}{\epsilon} \right)^{1-n} \quad (11)$$

What is important in Eq. (11) is the ϵ -dependence $(\epsilon_c/\epsilon)^{1-n}$ and not the constants multiplying it. In fact $P(\epsilon)$ is normalized to unity by the condition

$$\int_0^{\epsilon_c} P(\epsilon) d\epsilon = 1 \quad (12)$$

This readily implies that the normalization constant is n/ϵ_c and the form for $P(\epsilon)$ we shall henceforth consider is

$$P(\epsilon) = (n/\epsilon_c) (\epsilon_c/\epsilon)^{1-n} \quad (13)$$

Return to the sudden transitions of carriers in any conduction process, from state Φ_i to state Φ_j . If other elementary excitations (such as phonons in phonon-assisted hopping conductivity) are participating in addition to the charged particle, then the wave functions Φ 's is a composite wave function describing both the particle and the other elementary excitations. Had it been the case that no correlated states are excited, the end state of the transition, Φ_j , has the form of

$$\Phi_j = \psi_j e^{-iE_j t}$$

with ψ_j is the spatial part, and normalized to unity. In reality, accompanying the transition correlated state excitations are emitted. The probability amplitude $p(\epsilon)$ for the transition into Φ_j accompanied by correlation state excitations with energy ϵ can be deduced from the probability distribution $P(\epsilon)$ by taking the square root of $P(\epsilon)$. From Eq. (13),

$$p(\epsilon) = n^{1/2} (\epsilon/\epsilon_c)^{n/2} \epsilon^{-1/2} \quad (14)$$

The wave function after transitions is then of the form

$$\Phi_j = \int_0^{\epsilon_c} p(\epsilon) a_{\epsilon}^{\dagger} e^{i\epsilon t} \Phi_j d\epsilon e^{-iE_j t} \quad (15)$$

where a_{ϵ}^{\dagger} is the creation operator for a correlated state excitation of energy ϵ . For simplicity but without loss of generality in the subsequent development, we shall not consider de-excitation of correlated states in transition into Φ_j .

The elastic component of the probability amplitude of the wave function Φ_j is $p(\epsilon = 0)$, which, as seen in Eqs. (14-15) is drastically modified by infrared divergent response from a Dirac delta function to a weak singularity at $\epsilon = 0$. Although in principle the true elastic component is at $\epsilon = 0$, in actual practice any experimental setup to measure low energy or frequency will not have infinite resolution at the lowest frequencies. Hence there is a lower limit of energy ϵ_0 below which, the experimenter will not be able to distinguish an inelastic process (i.e. $\epsilon \neq 0$ in Eq. (15)) from an elastic process (i.e. $\epsilon = 0$). This suggests that the integral in Eq. (15) should be decomposed into two parts

$$\int_0^{\epsilon_c} = \int_0^{\epsilon_0} + \int_{\epsilon_0}^{\epsilon_c}$$

and identify the integral from 0 to ϵ_0 as the elastic part of the transition. The correct prescription is to integrate $P(\epsilon)$ from 0 to ϵ_0 , take the square root of this integral and interpret it as the elastic component probability amplitude. When this is carried out, the wave function takes the form of

$$\Phi_j = \left(\frac{\epsilon_0}{\epsilon_c}\right)^{n/2} \left\{ 1 + \int_{\epsilon_0}^{\epsilon_c} n^{1/2} \left(\frac{\epsilon}{\epsilon_0}\right)^{n/2} \epsilon^{-1/2} a_{\epsilon}^{\dagger} e^{i\epsilon t} d\epsilon \right\} \psi_j e^{-iE_j t} \quad (16)$$

To explain the phenomena of $1/f$ noise in a diverse number of materials and devices with many different conduction mechanisms would be an ambitious and most tedious task. We shall nevertheless attempt to do so to a certain extent. This is possible through an observation that the fundamental formula for conductivity due to Kubo²² and Greenwood²³ is quite versatile and can be suited up to describe several conduction mechanisms. For example, Edwards²⁴ has shown that it enables one to evaluate the conductivity in metals which correspond to scattering of electrons by weak-scattering potentials. Emin¹⁸ has started out with the Kubo-Greenwood formula and obtained the polaron hopping conductivity in molecular crystals. The Kubo-Greenwood formula has been given in several forms. The simplest and elementary version is,²⁴ for isotropic conductivity,

$$\sigma = -2\pi e^2 n \sum_{i,j} \langle \phi_i | v | \phi_j \rangle \langle \phi_j | v | \phi_i \rangle \delta(E_i - E_j) \frac{\partial f}{\partial E_i} \quad (17)$$

where $\langle \phi_i | v | \phi_j \rangle$ is the matrix element of velocity, and f is the electron distribution function. Quite often the Kubo-Greenwood formula is equivalently expressed in a form which relates the conductivity to a velocity-velocity correlation function or a current-current correlation function. It is in this alternate form that has been employed by Mahan²⁵ to calculate the ion hopping conductivity in ionic conductors such as crystalline salts AgI , CaF_2 and RbAg_4I_5 ; as well as by Fulde et al²¹ to describe Brownian particles mobility and conductivity. The Kubo-

Greenwood formula is also often the starting point of various possible conductivity considerations in disordered, noncrystalline solids.²⁶

Hence, from the discussions in the previous paragraph, our theory of 1/f noise which will start out from the Kubo-Greenwood formula Eq. (17) can support the claim that the same theory is indeed generally applicable to a diverse number of materials with different conduction mechanisms and carrier types (electrons or ions).

It is seen from Eq. (17) that the products $\langle \phi_i | v | \phi_j \rangle^* \langle \phi_i | v | \phi_j \rangle$ need to be considered for wave functions $\phi_i = \psi_i e^{-iE_i t}$ before the transition and ϕ_j as given in Eq. (16) after transition. Also, we have $E_i = E_j$ as required. It readily follows that

$$\sigma = -2\pi e^2 \hbar \sum_{i,j} \delta(E_i - E_j) \frac{\partial f}{\partial E_i} |\langle \psi_i | v | \psi_j \rangle|^2 (\epsilon_0 / \epsilon_c)^n \left\{ 1 + \int_{\epsilon_0}^{\epsilon_c} \epsilon^{-1/2} d\epsilon g(\epsilon) \right. \\ \left. (e^{i\epsilon t} a_{\epsilon}^{\dagger} + e^{-i\epsilon t} a_{\epsilon}) + \int_{\epsilon_0}^{\epsilon_c} \epsilon^{-1/2} d\epsilon \int_{\epsilon_0}^{\epsilon_c} \epsilon'^{-1/2} d\epsilon' g(\epsilon) g(\epsilon') e^{-i(\epsilon - \epsilon')t} a_{\epsilon} a_{\epsilon'}^{\dagger} \right\} \quad (18)$$

where the function $g(\epsilon)$ is

$$g(\epsilon) = n^{1/2} (\epsilon / \epsilon_0)^{n/2} \quad (19)$$

From the relation $J = \sigma \mathcal{E}$ which gives the current density J in terms of the product of the conductivity and electric field \mathcal{E} , and denoting the contribution to the current density from the i -th carrier as J_i , we have for unit \mathcal{E} ,

$$J = \sum_i J_i \quad (20)$$

$$J_i = -2\pi e^2 \hbar \sum_j \delta(E_i - E_j) \frac{\partial f}{\partial E_i} |\langle \psi_i | v | \psi_j \rangle|^2 (\epsilon_0 / \epsilon_c)^n \left\{ 1 + \int_{\epsilon_0}^{\epsilon_c} \epsilon^{-1/2} d\epsilon g(\epsilon) \right. \\ \left. (e^{i\epsilon t} a_{\epsilon}^{\dagger} + e^{-i\epsilon t} a_{\epsilon}) + \int_{\epsilon_0}^{\epsilon_c} \epsilon^{-1/2} d\epsilon \int_{\epsilon_0}^{\epsilon_c} \epsilon'^{-1/2} d\epsilon' g(\epsilon) g(\epsilon') \right. \\ \left. e^{-i(\epsilon - \epsilon')t} a_{\epsilon} a_{\epsilon'}^{\dagger} \right\} \quad (21)$$

The expectation value of J_i is

$$\langle J_i \rangle = -2\pi e^2 \hbar \sum_j \delta(E_i - E_j) \frac{\partial f}{\partial E_i} |\langle \psi_i | v | \psi_j \rangle|^2.$$

$$\cdot (\epsilon_0 / \epsilon_c)^n \left\{ 1 + \int_{\epsilon_0}^{\epsilon_c} g^2(\epsilon) d\epsilon / \epsilon \right\} \quad (22)$$

where we have used the fact that $\langle a_{\epsilon} a_{\epsilon'}^\dagger \rangle = \delta(\epsilon - \epsilon')$. It can be easily verified that

$$(\epsilon_0 / \epsilon_c)^n \left\{ 1 + \int_{\epsilon_0}^{\epsilon_c} g^2(\epsilon) d\epsilon / \epsilon \right\} = 1 \quad (23)$$

and hence $\langle J_i \rangle$ of Eq. (22) is independent of ϵ_0 , as it should.

The current fluctuation operator's δJ and δJ_i are defined by

$$\delta J = J - \langle J \rangle \quad (24)$$

$$\delta J_i = J_i - \langle J_i \rangle \quad (25)$$

with

$$\delta J = \sum_i \delta J_i \quad (26)$$

and it follows from Eqs. (21)-(22),

$$\begin{aligned} \delta J_i = & -2\pi e^2 \hbar \sum_j \delta(E_i - E_j) \left(\frac{\partial f}{\partial E_i} \right) |\langle \psi_i | v | \psi_j \rangle|^2 \cdot \\ & \cdot (\epsilon_0 / \epsilon_c)^n \left\{ \int_{\epsilon_0}^{\epsilon_c} \epsilon^{-1/2} d\epsilon g(\epsilon) (e^{i\epsilon t} a_{\epsilon}^\dagger + e^{-i\epsilon t} a_{\epsilon}) \right. \\ & - \int_{\epsilon_0}^{\epsilon_c} g^2(\epsilon) d\epsilon / \epsilon + \int_{\epsilon_0}^{\epsilon_c} \epsilon^{-1/2} d\epsilon \int_{\epsilon_0}^{\epsilon_c} \epsilon'^{-1/2} d\epsilon' g(\epsilon) \cdot \\ & \left. \cdot g(\epsilon') e^{-i(\epsilon - \epsilon')t} a_{\epsilon} a_{\epsilon'}^\dagger \right\} \end{aligned} \quad (27)$$

The autocorrelation function $C_i(\tau)$ in time τ of the current fluctuation

δJ_i is defined as conventionally¹⁻⁵ done by

$$C_i(\tau) = \frac{1}{2} \left\{ \langle \delta J_i(t + \tau) \delta J_i(t) \rangle + \text{its complex conjugate} \right\} \quad (28)$$

where the symbol $\langle \dots \rangle$ now denotes expectation value of the time

average (i.e. average over t). With δJ_i explicitly given as in Eq. (27), $C(\tau)$ can be readily evaluated. If n is small, from Eq. (19) it is clear that we need to keep the lowest order in $g(\epsilon)$ nonzero contribution to $C(\tau)$. Then, in this case, we have approximately

$$C_i(\tau) = \sigma_i^2 (\epsilon_o / \epsilon_c)^{2n} \int_{\epsilon_o}^{\epsilon_c} g^2(\epsilon) \cos \epsilon \tau \, d\epsilon / \epsilon \quad (29)$$

where now σ_i stands for the quantity

$$\sigma_i = -2\pi e^2 \hbar \sum_j \delta(E_i - E_j) \frac{\partial f}{\partial E_i} |\langle \psi_i | v | \psi_j \rangle|^2 \quad (30)$$

on the right hand side. Now the Wiener-Khintchine theorem¹⁻⁵ states that the power spectral density $G(f)$ defined as in Eq. (2) is related to the autocorrelation of the current density fluctuation

$$G_i(f) = 4 \int_0^\infty C_i(\tau) \cos(2\pi f \tau) \, d\tau \quad (31)$$

This relation together with our result for $C_i(\tau)$ as given in Eq. (29)

lead us to identify the current fluctuation power spectral function

$G_i(f)$ with the integrand in Eq. (29). This identification is justified

by recalling the definition of $\epsilon_o \equiv 2\pi f_o$ that $f_o \ll f$, and assuming

that $f \ll \epsilon_c / 2\pi \equiv f_c$ where f is the range of low frequency in which the

noise spectrum is $1/f$ like. Explicitly we have

$$G_i(f) = \sigma_i^2 (\epsilon_o / \epsilon_c)^{2n} \{ g(2\pi f) \}^2 f^{-1} \quad (32)$$

From Eq. (22), the ratio

$$\frac{G_i(f)}{\langle J_i \rangle^2} = n \left(\frac{f_o f}{f_c^2} \right)^n f^{-1} \quad (33)$$

has the frequency dependence

$$G_i(f) / \langle J_i \rangle^2 \propto f^{-\beta} \quad (34)$$

with

$$\beta = 1-n \quad (35)$$

Comparison of Eqs. (32-35) with Eq. (1) leads us to conclude that infrared divergent response of correlated states, that will always occur in any conduction process, causes their power spectrum of the current fluctuation to have the $1/f$ noise characteristics.

Consider now the totality of all the carriers, and, for simplicity, assumed here to be equivalent in the sense that they contribute equal current density J_i . If the total number of these equivalent carriers is N , then from Eq. (20),

$$\langle J \rangle^2 = \langle \sum_i J_i \rangle^2 = N^2 \langle J_i \rangle^2 \quad (36)$$

The power spectral function $G(f)$ for the total current fluctuation δJ is related to δJ by Eq. (2), hence we have from Eq. (26),

$$\int_0^\infty G(f) df = \langle (\sum_i J_i)^2 \rangle = \sum_i \langle (\delta J_i)^2 \rangle = \sum_i \int_0^\infty G_i(f) df \quad (37)$$

because $\langle \delta J_i \delta J_j \rangle = 0$ for $i \neq j$. Hence we have for N equivalent carriers,

$$G(f) = \sum_i G_i(f) = N G_i(f) \quad (38)$$

and

$$\frac{G(f)}{\langle J \rangle^2} = \frac{n}{N} \left(\frac{f_0 f}{f_c} \right)^n f^{-1} \quad (39)$$

This result demonstrates once more that infrared divergent responses of correlated states which are always concomitants of carriers transport give rise to current fluctuations a $f^{-\beta}$ power spectrum with β close to unity. It is interesting to note the similarity in form of our derived result of $G(f)/\langle J \rangle^2$ with the empirical form suggested by Hooge,^{4,27}

which is in our notations essentially

$$\frac{G(f)}{\langle J \rangle^2} = \left(\frac{\alpha}{N} \right) f^{-1} \quad (41)$$

Our value of α given by

$$\alpha = n(f_0 f / f_c^2)^n \quad (42)$$

is determined largely by the quantity n . According to Eq. (5), n is a measure of the degree of correlated state excitations emitted in their transient infrared divergent response to carrier transitions, and it should also be the quantity that governs the $1/\omega^{1-n}$ dielectric response of the same material. α is also dependent on the frequency f , the upper cut-off frequency f_c of the correlation state excitation energy, and the experimental resolution threshold frequency f_0 . If however,

$$n \log (f_0 f / f_c^2) \ll 1 \quad (43)$$

then the dependence of α on $(f_0 f / f_c^2)$ is weak and α to a good approximation is n . Starting with small n such that the inequality (43) is satisfied and assuming that f , f_0 and f_c are both fixed, it can be verified easily that α is an increasing function of n .

The derivation we have given for $1/f$ noise based on infrared divergent correlation states response is manifestly a mobility fluctuation theory. This is clearly seen from the relation

$$\mu = \sigma / Ne \quad (44)$$

between mobility μ and the conductivity σ , and the fact that we have based our conductivity on the Kubo-Greenwood formula and we have been considering for a fixed number of carriers N the current fluctuations. It being a mobility fluctuation theory worth further emphasis in view

of the fact⁴ that no model has been proposed for such mobility fluctuations.²⁸

IV. Discussions

One interesting and important characteristic of our theory on $1/f$ noise which is worth repeated emphasis is that it is a unified theory, explaining on the same basis both the ubiquitous $1/f$ noise and the universal $1/\omega^{1-n}$ low frequency dielectric response in current conducting condensed matter. As far as we know no existing theoretical model on $1/f$ noise does that, though in our view it should because dielectric loss and current fluctuations must be related in some fundamental way. Our theory traces the origin of both the $1/f$ noise and the $1/\omega^{1-n}$ dielectric response to energy structures (i.e. electronic energy structures as well as ionic or molecular interaction energy structures) and their excitations in condensed matter. In this sense, the theory goes deep into the structure of matter in identifying the origin of $1/f$ noise. These energy structures, states and their excitations are not the common elementary excitations in condensed matter, however. The effort in identifying them and their properties is the crux of the matter. Anyone who has some experience with the infrared divergence phenomena in Cerenkov radiation¹¹ or in the X-ray edge singularity of metals¹⁰ would be led to suspect some infrared divergent behavior may be operating in $1/f$ noise or in $1/\omega^{1-n}$ dielectric response. However, common elementary excitations in condensed matter such as phonons, electron-hole excitations etc., will not have infrared divergent response at finite temperature and at such low frequencies. Thus the task of providing the

source of low energy excitations for infrared divergent response is of paramount importance.

We have derived a mobility fluctuation theory of $1/f$ noise. The frequency dependence of the power spectrum of the mobility fluctuations whether the conductor is in equilibrium or carrying a direct current has the form of $1/f^{1-n}$, with n positive. The power spectrum Eq. (39) can be cast in a form closely resembles the empirical form Eq. (40) suggested by Hooge.^{4,27} The value of α , as can be seen in Eq. (41), depends on the infrared divergence of the correlated states through n and f_c . Both n and f_c depend on the energy structure of the correlated states which is determined by the interactions and correlations of the system. Therefore, it is not surprising in our model that one can have α to assume values which can differ by many orders of magnitude as experimentally observed. For examples, (a) in ionic conduction²⁹ it was found that α is 10^4 times larger than 2×10^{-3} , the value for α in bulk semiconductors and in metals;⁴ (b) in Ga doped Ge infrared detectors at cryogenic temperatures,³⁰ the value of α can be several orders of magnitude smaller; (c) the situation as in (b) has been reported for GaAs resistors;³¹ and (d) in very thin metal films where conduction is by hopping rather than by band conduction as in bulk metal,³² α is appreciably larger than 2×10^{-3} . It is too early to make quantitative prediction of α from our theory. Nevertheless, it is possible that these observed trends can be heuristically understood from the dependences of α on n and f_c (Eq. 42). For example, if we assume that the correlated states derived from ion-ion interactions to have appreciably smaller f_c than that derived from electronic interactions and correla-

tions then $\alpha_{ion} \gg \alpha_{el}$ as discussed in (a). Lower temperature may decrease the degree of excitation of the correlated states in the infrared divergent response (see Eq. (5)) and leads to a smaller value of n as well as a smaller value of α . Very thin metal films introduce discontinuities, boundaries, "interfaces" and as a consequence additional correlated state excitations than that in bulk films. This trend may also be correlated with (d). These arguments here are imprecise and may be oversimplified. They are given here not for the purpose of lending support to our model but rather employed as vehicles to discuss the dependences of α in our model.

In closing we mention that the number or concentration of charged carriers fluctuations induced low frequency noise have also been examined by us in the infrared divergent response formulation. Concentration fluctuations of charged carriers can be most clearly visualized in conduction process with the presence of carrier trapping centers. Trappings of carriers as well as detrappings of carriers at these centers not only cause a change in the number of free carriers present as often been observed, but also switch on or switch off a new potential for the correlated states to respond as a center is being filled or being emptied. Again like in the earlier mobility fluctuation theory where carrier transitions occur in times much shorter than both the characteristic times of the correlated states and $1/2\pi f$, we also assume the same for the trapping and detrapping time constants. Hence the new potential switched on or switched off is sudden for the correlated states to respond, and all the conditions for infrared divergent response including Eq. (5) are therefore satisfied. We have carried

out the analysis of the autocorrelation function of the concentration fluctuations. For concentration fluctuation ΔN associated with a trapping center with time constant τ_0 , the autocorrelation function is modified from the familiar form of²

$$C(t) = \exp(-t/\tau_0) \quad (45)$$

to a drastically different form of

$$C(t) = e^{-n\gamma} (\epsilon_c t)^{-n} \exp(-e^{-n\gamma} t^{1-n} / (1-n) \tau_0 E_c^n) \quad (46)$$

with the symbols as defined in Eqs. (5) and (11). When the infrared divergent excitations of the correlated states are taken into consideration, one may expect the noise power spectrum to be modified. The preliminary conclusions we arrive at are that concentration fluctuation in the presence of infrared divergent response does not give an $1/f$ noise spectrum unless one assumes a wide range of time constants τ with some chosen statistical weight such as inversely proportionality to τ .² However, even with one time constant we find it is possible to have a $1/f^{1-n}$ spectrum, with n less than unity but not small, over an extended frequency interval which flattens off at low frequencies. Details of these investigations into concentration fluctuation and implications of their results will appear elsewhere.

V. Summary

We have presented a theoretical model of mobility fluctuations that has a $1/f$ power spectra in electric current conducting materials. As far as we know this is the first mobility fluctuation theory of $1/f$ noise that is general enough to cover various types of current conduction in condensed matter. The model applies to both electronic and

ionic conductions, as well as to both bulk and interface conductions. The present theory traces the origin of the $1/f$ noise to some electronic or ionic energy structure (called correlated states in this work) and excitations. These low energy excitations of the correlated states exhibit an infrared divergent behavior and give rise to both the ubiquitous $1/f$ noise and the "universal" dielectric response of condensed matter. It is in this sense that the present work is a unified theory, explaining two universal phenomena with a single physical picture.

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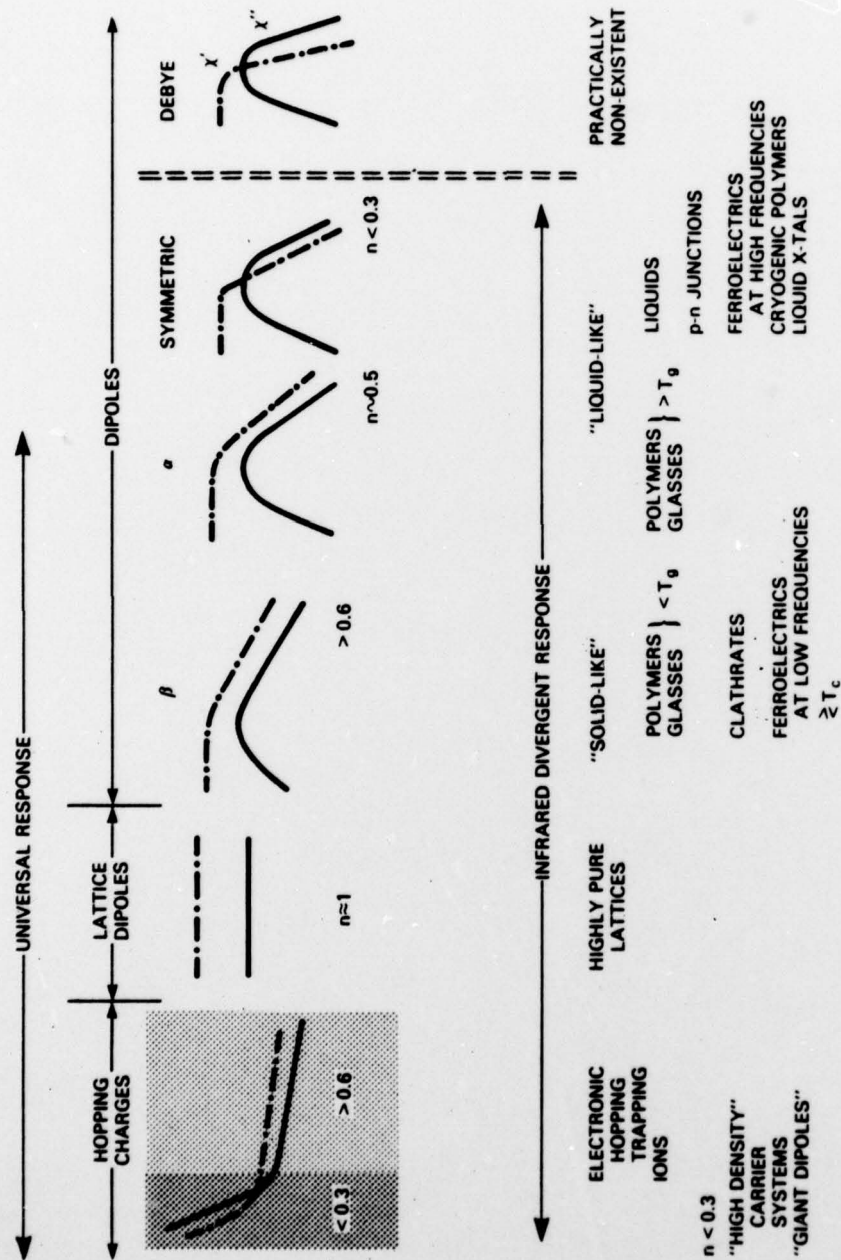


Fig. 1 — A schematic representation of the various observed types of dielectric response in the entire range of solids. The set of diagrams represents the shapes of the logarithmic plots of the real part of the dielectric function $\epsilon'(\omega)$ - chain - dotted lines, and the imaginary part $\epsilon''(\omega)$ - solid lines, ranging from the ideal Debye through the α and β peaks for dipolar dielectrics and on to the universal $1/\omega^{1-n}$ dependence for charged carrier systems (shaded portion). The limiting forms of behavior are represented by the strong low-frequency dispersion with small values of n (darkly shaded portion) for charged carrier systems. The other type of behavior with nearly frequency-independent ϵ' and ϵ'' labelled as "lattice response" with $n \approx 1$ is also illustrated. Examples of the various types of materials obeying the respective types of response are shown. The present work considers only the shaded portion of the behaviors for materials with charged carriers.